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ADVANCED EPITAXIAL FERRITE DEVICES

PROJECT NO. IT 161102BH57-03

FINAL REPORT

W. L. BONGIANNI

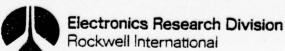
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It was the purpose of this contract to explore the use of epitaxial films of hexagonal ferrites as an alternative to bulk grown single crystal material.

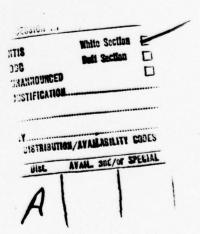
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Since epitaxial films can be grown with a damage-free surface, no mechanical handling would be required.

Bandstop filters using this material were fabricated and the characteristics were measured. Using parallel resonance a bandstop response at 39.1 GHz was obtained with an unloaded Q of 400. In perpendicular resonance, the film was linearly tuned across the 50 to 60 GHz range with a variable field which did not exceed 2000 gauss. In both cases only a minimum of mechanical processing was used.



The findings in this report are not to be construed as an official Department of the Army position, unless so designated by other authorized documents.

# TABLE OF CONTENTS

		_	Page No.
I.	INTRODUCTION	•	1
II.	RESONANCE OF EPITAXIAL SINGLE-CRYSTAL HEXAGONAL FERRITES		1
III.	MATERIAL PREPARATION AND CHARACTERIZATION		4
IV.	MICROWAVE CHARACTERIZATION		8
	A. Resonance Measurements		8
	B. Bandstop Characteristics and Linewidth		11
v.	CONCLUSIONS		15
VI.	PARTICIPATING SCIENTIFIC PERSONNEL		16
TII.	BIBLIOGRAPHY		16

#### I. INTRODUCTION

Using a class of materials referred to as hexagonal ferrites, magnetically tunable filters in the millimeter wave range becomes a practical possibility. Because of their large inherent magnetic anisotropy, the hexaferrites can be biased into the millimeter range with relatively small external fields.

One drawback with this material occurs when attempts are made to fabricate a sphere resonator from the raw bulk single crystal. Along with magnetic anisotropy comes an anisotropy of hardness which makes the attainment of a perfect sphere with a good surface finish exceedingly difficult. It was the purpose of this contract to explore the use of epitaxial films of hexagonal ferrites as an alternative to bulk single crystal material. Since epitaxial films can be grown with a damage—free surface, and since they can be etched chemically, no mechanical handling would be required.

During this contract, the initial steps to developing epitaxial hexaferrite filters were made. These steps consisted of identifying and growing potential single crystal substrates, growing epitaxial films, and testing the film in a bandstop filter configuration. Finally, a very narrow bandstop response was measured on one of the films in parallel resonance. This response appeared to be almost independent of applied field and had an unloaded Q of 400 at a frequency of 39.1 GHz.

# II. RESONANCE OF EPITAXIAL SINGLE-CRYSTAL HEXAGONAL FERRITES

The films considered for this program were M-type (BaFe<sub>12</sub>0<sub>19</sub>) hexagonal ferrites. Epitaxial growth gave a film with the hexagonal easy or c-axis normal to the plane of the film. Experimentally, the plane of the film is

easily identified; and therefore, parallel and perpendicular resonance refers to the application of the applied field parallel and perpendicular to the plane of the film respectively.

The resonance frequency for a uniaxial crystal is given by Buffler as:

(a) Uniaxial crystal with applied field in hard plane.

$$\left[\frac{\omega}{\gamma}\right]^{2} = \left\{1 - \frac{(N_{y} - N_{z}) M}{H_{a} - (N_{z} - N_{y}) M}\right\} \left\{\left[H_{a} - (N_{z} - N_{y}) M\right]^{2} - H_{o}^{2}\right\},\tag{1}$$

for 
$$H_0 \le H_a - (N_z - N_y)$$
 M and  $H_a > 4\pi M$ ;

$$\left[\frac{\omega}{\gamma}\right]^{2} = \{H_{o} - (N_{y} - N_{z}) M\} \{H_{o} - [H_{a} - (N_{z} - N_{y}) M]\}$$
 (2)

for 
$$H_0 \ge H_a - (N_z - N_y)$$
 M and  $H_a > 4\pi M$ 

In this case the hexagonal easy or c-axis is taken as the z direction and the applied field  $H_{\rm o}$  is taken along the y-axis.

(b) Uniaxial crystal with applied field along easy axis.

$$\left[\frac{\omega}{\gamma}\right]^{2} = \left[ (H_{o} + H_{a}) - (N_{z} - N_{x}) M \right] \left[ (H_{o} + H_{a}) - (N_{z} - N_{y}) M \right]. \tag{3}$$

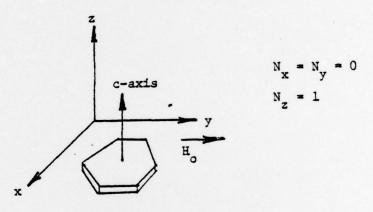
In this case the c-axis and the applied field are both taken along the z-axis. Where  $N_x$ ,  $N_y$ , and  $N_z$  are the demagnetization coefficients in the x, y, and z direction;

M is the saturation magnetization;

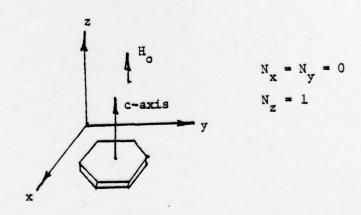
 $\mathbf{H}_{\mathbf{a}}$  is the magnetic anisotropy along the c-axis; and

H is the applied magnetic field.

Since the epitaxial film grows with the c-axis or easy direction normal to the film, two cases of parallel and perpendicular resonance occur as indicated in Figure 1.



(a) Parallel to the film.



(b) Perpendicular to the film.

Figure 1. Resonant geometry of a uniaxial film with a normal c-axis.

The two geometries give:

(a) Parallel resonance.

$$\left(\frac{\omega}{\gamma}\right)^2 = (H_a - 4\pi M)^2 - H_o^2$$
  
for  $H_o \le H_a - 4\pi M$  and  $H_a > 4\pi M$ ;

$$\left(\frac{\omega}{\gamma}\right)^{2} = H_{o}(H_{o} - H_{a} + 4\pi M)$$
for  $H_{o} \ge H_{a} - 4\pi M$  and  $H_{a} > 4\pi M$ . (5)

(b) Perpendicular resonance.

$$\left(\frac{\omega}{\gamma}\right) = H_0 + H_a - 4\pi M. \tag{6}$$

Epitaxial films were grown on a number of different substrates. The substrates were flux grown single crystals, which measured at most a few millimeters in diameter. Since this represented too small an area to chemically etch a disk from, sample preparation consisted of choosing the best available face and cutting out a plate with the face as one side. Film thicknesses were about 1 mil in all cases measured.

#### III. MATERIAL PREPARATION AND CHARACTERIZATION

The hexagonal ferrite samples used in this study were grown in our laboratory as part of a research contract with the Air Force Office of Scientific Research, F44620-75-C-0045 "Magnetostatic Surface Wave Materials." The LPE (liquid phase epitaxy) dipping method was employed to deposit the hexagonal ferrites on various single crystal substrates. Details of the LPE method have been published<sup>2,3</sup>. It should be borne in mind that the LPE method itself and the preparation of suitable substrates are still in

the research stage. Rapid progress is being made in these research efforts, but the materials reported on here are far from optimum in quality.

Although we have been able to grow several different hexagonal ferrite types by LPE, the samples used in this study were all of the M-type, BaFe<sub>12</sub>0<sub>19</sub>, which is well suited to the frequency range of interest. Hexagonal ferrite type is determined from x-ray diffractograms of multiple orders of basal plane reflections. The intensities and angular positions of the various orders are characteristic of the type. Non-basal reflections may be used to establish the epitaxial (orientation) relationships between films and substrates when they are not obvious from morphology.

Part of the AFOSR program involves a study of the relation between

ZnO content of the LPE melt and hexagonal ferrite type of the deposit. The

samples described in this report were grown from a melt which contained ZnO,

but at a concentration which was low enough that the deposits were M-type.

Properly, these samples should be considered as Zn-doped. The doping level

was low since the M-type ferrite cannot accommodate much Zn unless an

additional dopant is present to provide charge-compensation. It is possible,

however that the Zn content was sufficient to have produced perceptible

shifts in some magnetic parameters.

The two samples most intensively studied, BFO-9L-40 and BFO-9L-42, were grown from the same LPE melt but on different substrates. Sample BFO-9L-40 was deposited on an octahedron-shaped single crystal of  ${\rm ZnGa_2O_4}$ , a non-magnetic spinel. This substrate was grown from a flux by the conventional slow-cooling method as part of our in-house IR&D program. The natural (III) facets of the spinel are of the proper orientation for epitaxial growth of the hexagonal ferrite, so the entire  ${\rm ZnGa_2O_4}$  crystal was used as a substrate for LPE growth without any further preparation.

The hexagonal ferrite layer was deposited at a temperature of 935°C for 58 minutes. The layer, which covered all of the substrate except for a small area which was not of the correct (III) orientation, was estimated to be between 10 and 20µm thick.

X-ray diffraction of BFO-9L-40 confirmed that the layer was M-type and single clystal with basal planes parallel to the substrate (111). The azmuthal orientation relation could not be determined directly by x-ray diffraction because the layer was too thick. Measurements of layers on other spinel substrates have shown that the hexagonal ferrite a-axis is parallel to the spinel [101] direction.

The x-ray technique only searched for hexagonal ferrite with the expected orientation; i.e., with basal plane nearly parallel to the substrate surface. Scanning electron microscopy showed that the layer contained areas with hexagonal-shaped platelets standing nearly on edge rather than lying parallel to the substrate. These "upright" platelets, Figure 2, were not randomly oriented but were predominantly aligned with their basal planes parallel to other {111} substrate planes. Thus, the "upright" platelets are also epitaxial. Their formation may have been related to defects on the substrate surface. While the sample was in the scanning electron microscope, an energy dispersive x-ray spectrochemical analysis was performed. The only elements detected in large concentrations were Fe and Ba (elements with atomic number less than 10 are not detectable with this system.) Traces of Zn and Ga were also found. It was not clear whether the Zn and Ga signals arose from the substrate or the deposit. If Zn and Ga were in the deposit, their concentrations were less than 1% that of Fe. In either case, the Zn/Fe ratio in the deposit was far less than that in the LPE melt, as expected.



Figure 2. Scanning electron micrograph of sample BEO-9L-40 at 650X magnification.

The substrate for sample BFO-9L-42 was one of several crystals of Zn, Ti, Mn-doped M-type hexagonal ferrite supplied by A. Tauber,

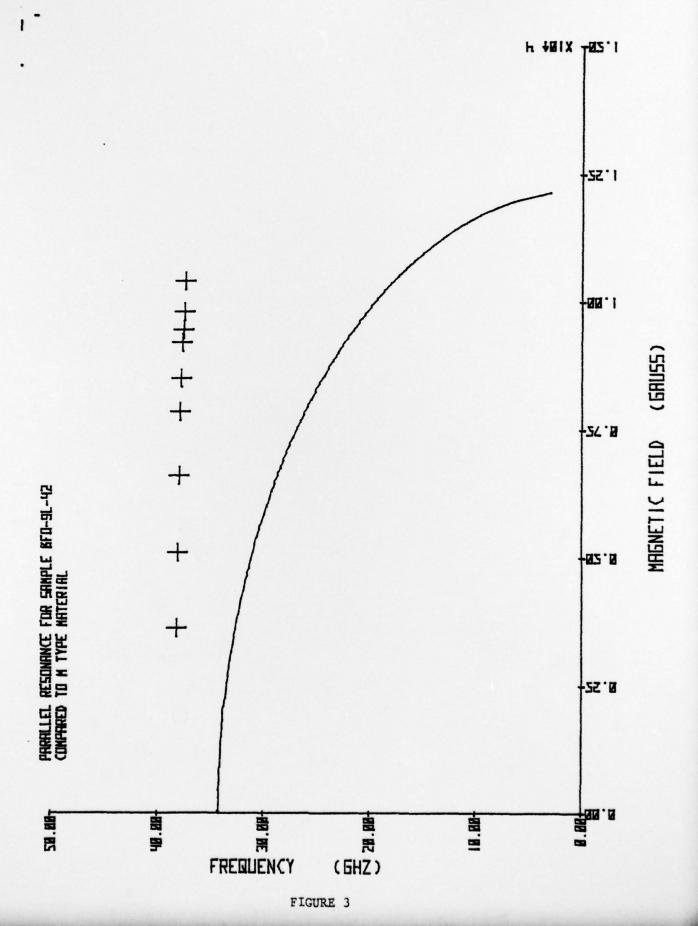
T. R. AuCoin and R. O. Savage of ECOM. The doping levels were sufficient to depress the Curie temperature below room temperature so the substrates may be considered non-magnetic. A hexagonal ferrite layer ~40µm thick was deposited on this substrate at 938°C for 283 minutes. The layer was mostly epitaxial; but some randomly oriented growth was observed, probably associated with surface defects. No x-ray analysis was performed; however, all previous deposits from this LPE melt were found to be M-type. Uniaxial magnetization along the c-axis was confirmed.

# IV. MICROWAVE CHARACTERIZATION

#### A. Resonance Measurements

Room temperature measurements were made using a bandstop filter configuration. This technique consists of placing the sample in the waveguide and performing a transmission measurement; that is, holding the sample at a fixed magnetic bias, the frequency is swept and any loss of transmitted energy is measured. Figure 3 is the plot of the parallel resonance frequency measured for sample BFO-9L-42. In this sample resonance occurred at 38 GHz with very little frequency shift as a function of field - the frequency shifted from 38.2 GHz at 3.62 kG to 37.4 GHz at 10.40 kG. The theoretical resonance for an M type material is shown in Figure 3 for comparison. While there is a downward trend to the data, it does not follow the theory as given by equation (4). While this trend is not understood, it should not be completely unexpected as the sample is not saturated (H<sub>O</sub><H<sub>a</sub>-4πM).

Figure 4 is a plot of sample BFO-9L-40. In this sample, both parallel and perpendicular resonance were observed. As in the previous sample, parallel data was flatter than predicted. The perpendicular resonance is linear and ranges across the 50 to 60 GHz band for fields under 2 kG.



FREQUENCY (GHZ)

58.88

78.88

EB. 88

THE BE

-BB. BE

28.88

H. H.

### B. Bandstop Characteristics and Linewidth

The performance of a filter is normally given in terms of the quality or Q. Since this is made up of two components — the intrinsic loss of the resonator, given by the unloaded Q,  $Q_u$ , and the loss associated with the coupling circuit, given by the external Q,  $Q_e$ . The overall measured filter performance is given by the loaded Q or  $Q_1$ 

The  $Q_{\rm e}$  for a number of electromagnetic structures has been calculated by Carter  $^4$  for the case of ferrimagnetic resonators. Figure 5 is a sketch of the TE $_{10}$  structure used in most of the work reported here.

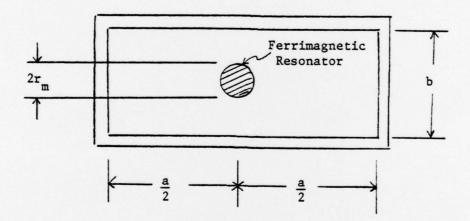


Figure 5. TE<sub>10</sub> Waveguide

If the ferrimagnetic resonator consists of a thin disk, the  $\mathbf{Q}_{\mathbf{e}}$  is given by

$$Q_{e} = \frac{60 \text{ ab}}{\mu_{o} \omega_{m} t r_{m}^{2}} (\frac{\lambda}{\lambda g})$$

where  $\omega_m = \mu_0 r (4\pi M)$ ,

 $\mu_{o}$  = intrinsic permeability of free space

=  $1.256 \times 10^{-6}$  henries per meter,

g = Lande "g" factor = 2.00 for electrons in most ferrites,

 $\gamma$  = gyromagnetic ratio = g(e/2m),

e/m = ratio of charge e to mass m of electron

= 1.759 x 10<sup>11</sup> coulombs per kg.

a,b = width and height of waveguide in inches,

t = film thickness in inches,

2r = disk diameter in inches,

 $\lambda$  = free space wavelength,

 $\lambda_g$  = guide wavelength.

A 100 mil diameter disk of 1 mil thickness would provide a  $Q_e = 630$  in  $K_a$  band waveguide at 35 GHz.

The unloaded Q is found by differentiating equations (4) and (6) and substituting into the definition  $Q_{ij} = f/\Delta f$  as:

(a) Parallel resonance.

$$Q_{u} = [(H_{a} - 4\pi M)^{2} - H_{o}^{2}]^{1/2} (H_{a} - 4\pi M)/(\Delta H \cdot H_{o}),$$
 (8)

(b) Perpendicular resonance.

$$Q_{u} = (H_{o} + H_{a} - 4\pi M)/\Delta H$$
 (9)

where H is the material linewidth.

The best response for a single resonator bandstop filter was obtained with sample BFO-9L-40. Figure 6 shows the response of this filter in parallel resonance. The analysis of a single resonator bandstop filter has been considered by Young et al for the simple circuit mode of Figure 7. In this case the unloaded Q is given by

$$Q_{u} = \frac{1}{\omega_{L_{A}-db}} \sqrt{\frac{A^{2} - 10^{L}A^{10}}{10^{L}A^{10} - 1}} . \tag{10}$$

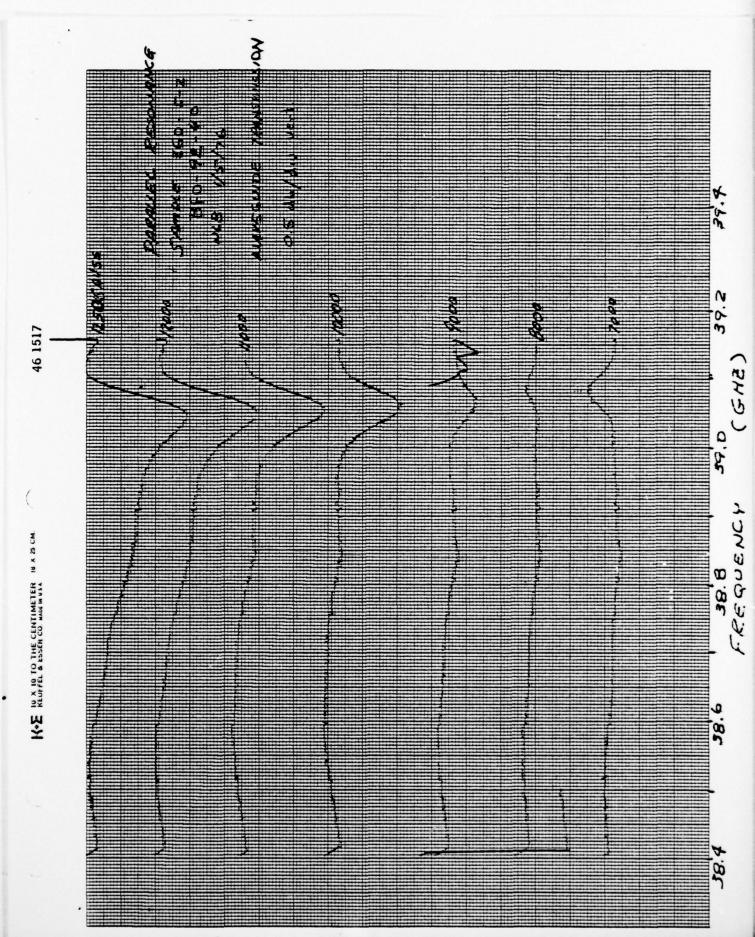


FIGURE 6

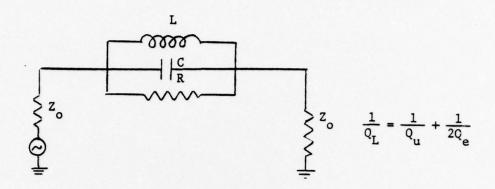


Figure 7. Equivalent Circuit for a Single Resonator Bandstop Filter.

where

 $(L_{A})_{max}$  = maximum attenuation in dB of bandstop filter (with single resonator).

ω<sub>x-dB</sub> = fractional bandwidth between x-dB points on a guide wave-length basis (It sπouId be emphasized that this is the bandwidth of a single cavity of possible a multi-cavity filter in which the other cavities have been decoupled.)

A = antilog<sub>10</sub> 
$$[\frac{(L_A)_{max}}{20}] = 10(L_A)_{max}/20$$
.

Using equation (10) and the data provided by Figure 6 yields, at 12,000 Gauss,  $Q_u = 400$ . The intrinsic linewidth of the material would generally be derived from equation (8), but the experimental results do not appear to follow the predicted values from which equation (8) is derived.

Instead, the linewidth can be estimated from the data and the unloaded Q. At 12 kG, the slope  $\Delta f/\Delta H = 0.008$  GHz/k Oe. Thus:

$$\Delta H = \frac{\Delta f}{.008} = \frac{\Delta f}{f} \cdot \frac{f}{.008} = \frac{f}{Q_{11}} (\frac{1}{.008}) \approx 12 \text{ k Oe.}$$

This exceedingly broad linewidth implies that the response of the filter is essentially independent of the bias field, which is supported by the data shown in Figure 6. It is also consistent with the fact that the sample is not saturated.

For perpendicular resonance, the unloaded Q measured 54 on sample BFO-9L-40. Equation (9) can be used to estimate the linewidth, since the behavior of the data conforms to the predicted slope.

Hence,

$$\Delta H = (H_0 + H_a - 4\pi M)/Q_u$$
 (11)

The results for the samples studied are given in Table 1.

Table 1

SAMPLE	RESONANCE	Q <sub>u</sub>	Н	FREQ. MEAS.
BF0-9L-40	Parallel	400	12K 0e	39.1 GHz
	Perpendicular	54	240 0e	53.0 GHz
BFO-9L-42	Parallel	55	3.2K 0e	37.55GHz
	Perpendicular	-	_	-

### V. CONCLUSIONS

Epitaxial hexagonal ferrite films can be used to produce fixed tuned and magnetically tunable bandstop filters. Using parallel resonance a bandstop response at 39.1 GHz was obtained with an unloaded Q of 400. In perpendicular resonance, the film was linearly tuned across the 50 to 60 GHz range with a variable field which did not exceed 2000 gauss. In both cases only a minimum of mechanical processing was used.

The linewidths were relatively poor. Since the state of the art for film and substrate growth is only a few months old, this is not surprising. With a reasonable amount of effort, film quality should improve to a considerable extent. And only a five fold improvement is needed for device quality material.

Although it is somewhat early to come to any major conclusions with respect to the theory of resonance, some modifications would be indicated if future films behave as do the two so far measured. In particular, parallel resonance does not appear to conform to theory in a straight forward fashion. It is obvious that resonance is occurring at fields below magnetic saturation. It thus appears that the assumption of magnetization along the c-axis cannot be maintained. This in turn implies a more detailed theoretical model which includes the effect of the domain wall energy and its effect on off axis resonance and linewidth.

#### VI. PARTICIPATING SCIENTIFIC PERSONNEL

The major activities of this program were performed by
Wayne L. Bongianni. Material support and film growth was by Dr.
Howard Glass. Program coordination was provided by Mr. Jerry C. Aukland.
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